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ANOMALOUS PRESSURE DEPENDENCE OF DIPOLAR RELAXATION  
TIMES IN RARE EARTH DOPED LEAD FLUORIDE(U) NAVAL  
ACADEMY ANNAPOLIS MD DEPT OF PHYSICS  
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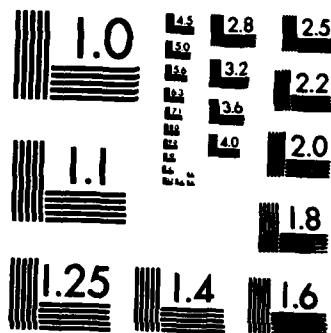
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ANOMALOUS PRESSURE DEPENDENCE OF DIPOLAR RELAXATION TIMES IN RARE  
EARTH DOPED LEAD FLUORIDE

by

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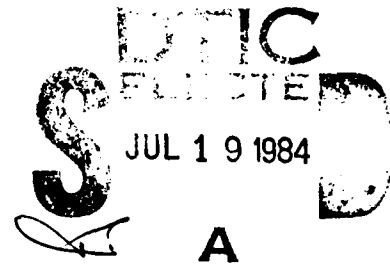
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| 20. ABSTRACT (Continue on reverse side if necessary and identify by block number)<br>Electrical relaxation measurements at high pressures have been carried<br>out on lead fluoride doped with lanthanum and cerium. A single, strong<br>relaxation peak is observed and the relaxation time decreases with pressure.<br>This is contrary to the behavior exhibited by all known relaxations and can<br>be explained by attributing the relaxation to a substitutional trivalent rare<br>earth compensated either by an interstitial fluorine ion with a soft attempt<br>mode or by an electron. |                       |  |

# Anomalous Pressure Dependence of Dipolar Relaxation

## Times in Rare Earth Doped Lead Fluoride

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### ABSTRACT

Electrical relaxation measurements at high pressures have been carried out on lead fluoride doped with lanthanum and cerium. A single, strong relaxation peak is observed and the relaxation time decreases with increasing pressure. This is contrary to the behavior exhibited by all known relaxations and can be explained by attributing the relaxation to a substitutional trivalent rare earth compensated either by an interstitial fluorine ion with a soft attempt mode or by an electron.



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will be discussed in detail elsewhere. For  $\text{PbF}_2\text{:Ce}$ , no conductivity correction was made and the results are quite similar to figure 2. The Cole-Cole expression was fitted to the data in order to determine the relaxation times at various pressures. The resultant relaxation times vs. pressure are shown in figure 3 for  $\text{PbF}_2\text{:La}$  and the slopes of the best fit straight lines for all samples are listed in Table 1. The decrease of relaxation time as pressure increases is opposite from that observed for any dipolar relaxation time studied to date including work on ionic crystals<sup>17-20</sup> or polymers.<sup>21</sup>

It may be that the anomalous pressure dependence of the relaxation time is another manifestation of the soft mode behavior which lead fluoride has been suggested to exhibit.<sup>1,5</sup> Specifically, it might be concluded that the usual reorientation of a charge compensating fluorine ion (IC) near a substitutional rare earth is taking place, and that the vibrational mode governing the reorientation is a soft mode. Possibly the best way to illustrate this is via the activation volume:

$$\Delta V_{\text{exp}}^* = kT \frac{\partial \ln \tau}{\partial P}$$

which yields the negative values listed in Table 1. The dynamical diffusion theoretical expression<sup>20</sup>

$$\Delta V_{\text{th}}^* = 2 \gamma_a X_T g$$

where  $X_T$  is the compressibility and  $g$  is the Gibbs energy, then requires a negative attempt mode Gruneisen parameter,  $\gamma_a$ , indicating a soft mode, to explain the data.

However, these relaxations exhibit a very strong dependence of peak position or activation energy on dopant ion size as is evident from Table 1 for  $\text{PbF}_2\text{:La}$  and  $\text{PbF}_2\text{:Ce}$  and figure 1 of reference 9. It may be that the same spring constants which soften upon application of pressure also enhance the interaction between the RE and the reorienting fluorine interstitial. IC has the additional advantage of easily explaining the relatively high activation energies, 0.4 and 0.32 eV, observed for La and Ce. However, the activation energy for the analogous relaxation in  $\text{PbF}_2\text{:Gd}$  is very low, about 0.11 eV. This suggests an alternative interpretation of the data. The second model proposed is that the RE is charge compensated by an electron (EC) localized around one of the adjacent lattice fluorine ions. The decrease in the relaxation time with increased pressure would then be a result of increased electron wavefunction overlap between equivalent wells as pressure reduces the lattice parameter. Further, these same overlap effects will make the electronic relaxation time very sensitive to the extent of the RE's electron wavefunction and thus explain the variation of activation energy with ion size.

These models have the following implications concerning several other unusual features of the electrical response of RE doped  $\text{PbF}_2$ .

Firstly, the magnitude of the conductivity does not scale with the strength of the principal relaxation over the dopant range 0.1 to 1.0 mole% (nominal) in that the strength of the relaxation increases while the magnitude of the conductivity does not. This result favors the EC model since there should be no

correlation of the strength of the relaxation and the magnitude of the conductivity which is primarily ionic.<sup>22,23</sup> (There have been reports of electronic contributions to the conductivity of lead fluoride<sup>24,25</sup> and this increases the plausibility of EC.) Of course, the IC model could explain the results if the association energy is so large that it is not the complex responsible for the extrinsic conductivity. In either case, another defect site must coexist with that responsible for the principal relaxation since the conductivity is well-behaved and can be fit via a classical association model.<sup>22,23</sup> In fact, a relatively weak relaxation is observed in both  $\text{PbF}_2\text{:La}$  and  $\text{PbF}_2\text{:Ce}$  at a slightly higher temperature than the principal relaxation. That is part of the reason for the poor fit of the data shown in figure 1. If that site has only a very small dipole moment the concentration could be sufficiently large to give rise to the extrinsic conductivity.

Secondly, the deviation of the relaxations from Curie-Weiss behavior can be understood either in terms of EC or IC. In the case of EC, it could easily be that the dipole moment increases as temperature increases. For either EC or IC a slight potential well inequivalency could also explain the trend.

Finally, there is the phenomenon of the existence of multiple low temperature relaxations per defect center observed for  $\text{PbF}_2$  doped with RE's smaller than gadolinium.<sup>8,9</sup> This has not been observed in association with ionic relaxations in the AEF. Consequently, those results suggest the existence of at least some electronic compensation.

In summary, it has been shown that for the single, strong



relaxation observed in  $\text{PbF}_2$  doped with large RE's, the relaxation time decreases as pressure increases. By attributing the relaxation to an electronically compensated or soft mode related, fluorine interstitial compensated substitutional trivalent RE, this anomalous behavior, together with some previously unexplained phenomena, can be understood.

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Table 1. Parameters describing the principal relaxation in 0.1 mol%  
PbF<sub>2</sub>:La and PbF<sub>2</sub>:Ce.

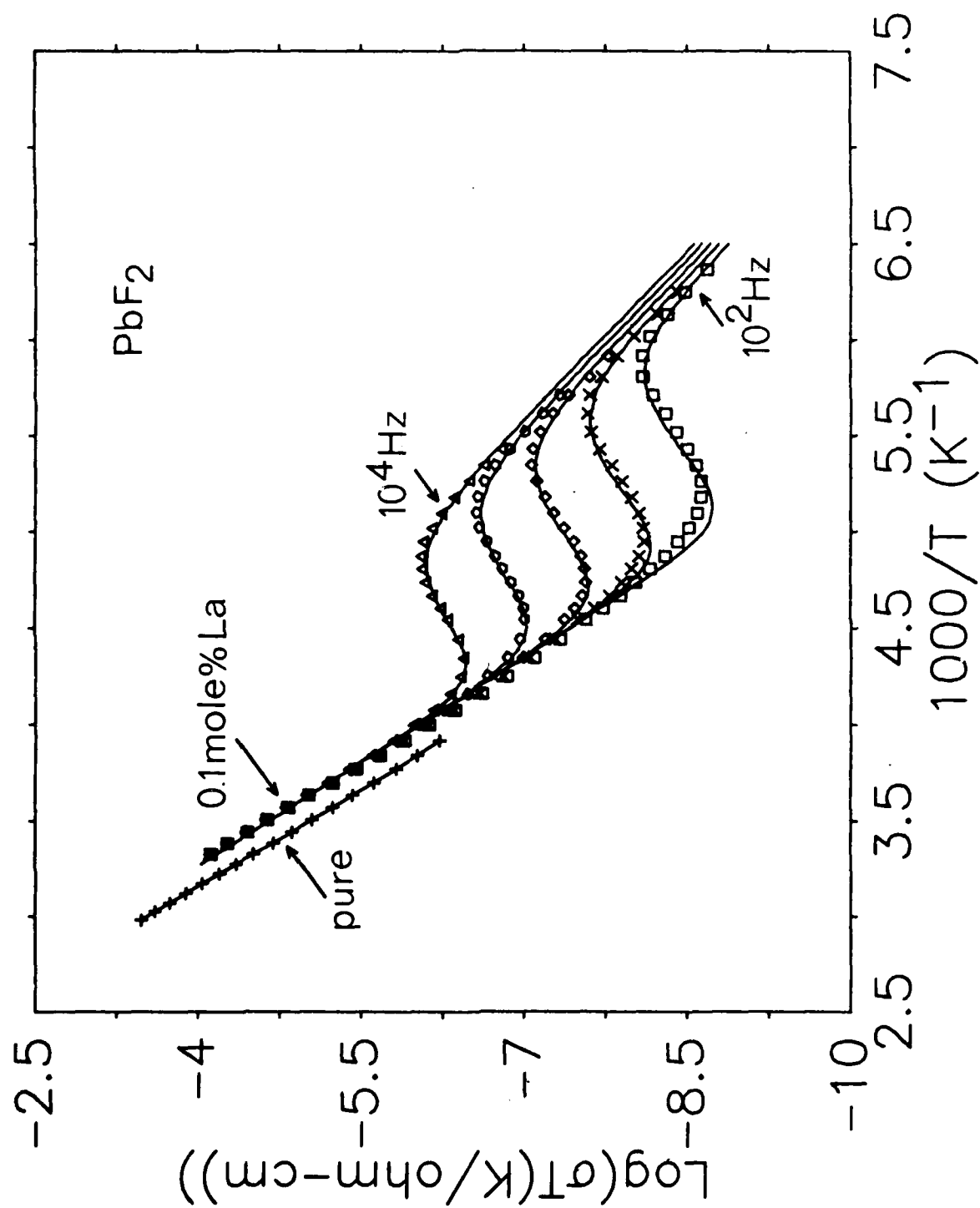
| Dopant | A(K) | $\tau_o (10^{-14} \text{ s})$ | E(eV) | $\alpha$ |          |      | $\frac{d \ln \tau}{dP} (\text{GPa}^{-1})$ | $\Delta V^* (\text{cm}^3/\text{mole})$ |
|--------|------|-------------------------------|-------|----------|----------|------|---|--|
| La     | 364  | 0.25                          | 0.40  | 0.14     | Sample 1 | 181K | -1.25                                     | -1.88                                  |
|        |      |                               |       |          |          | 185K | -1.23                                     | -1.89                                  |
|        |      |                               |       |          | Sample 2 | 181K | -1.17                                     | -1.76                                  |
|        |      |                               |       |          |          | 185K | -1.16                                     | -1.78                                  |
| Ce     | 261  | 1.6                           | 0.32  | 0.04     | Sample 1 | 161K | -1.34                                     | -1.79                                  |

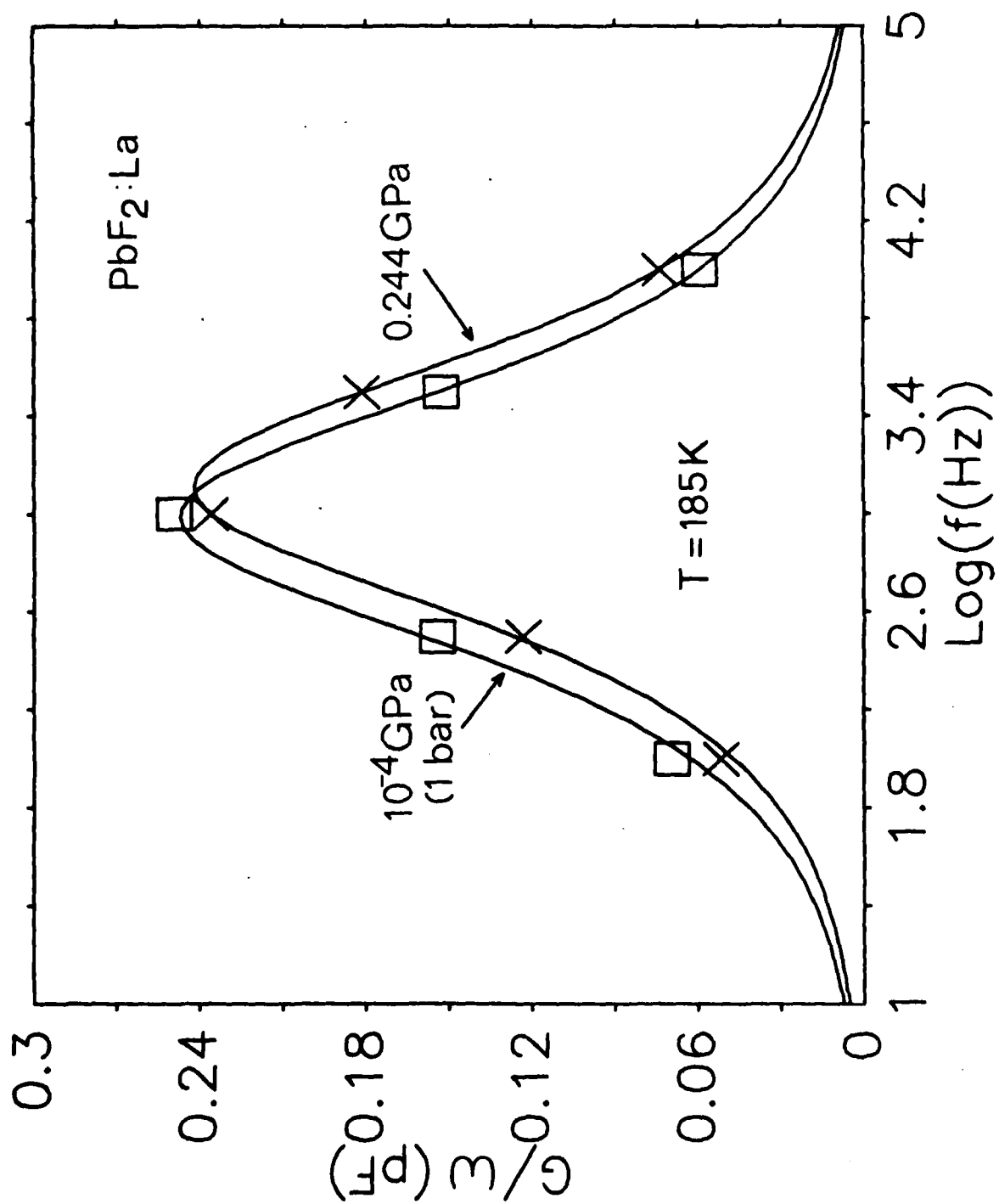
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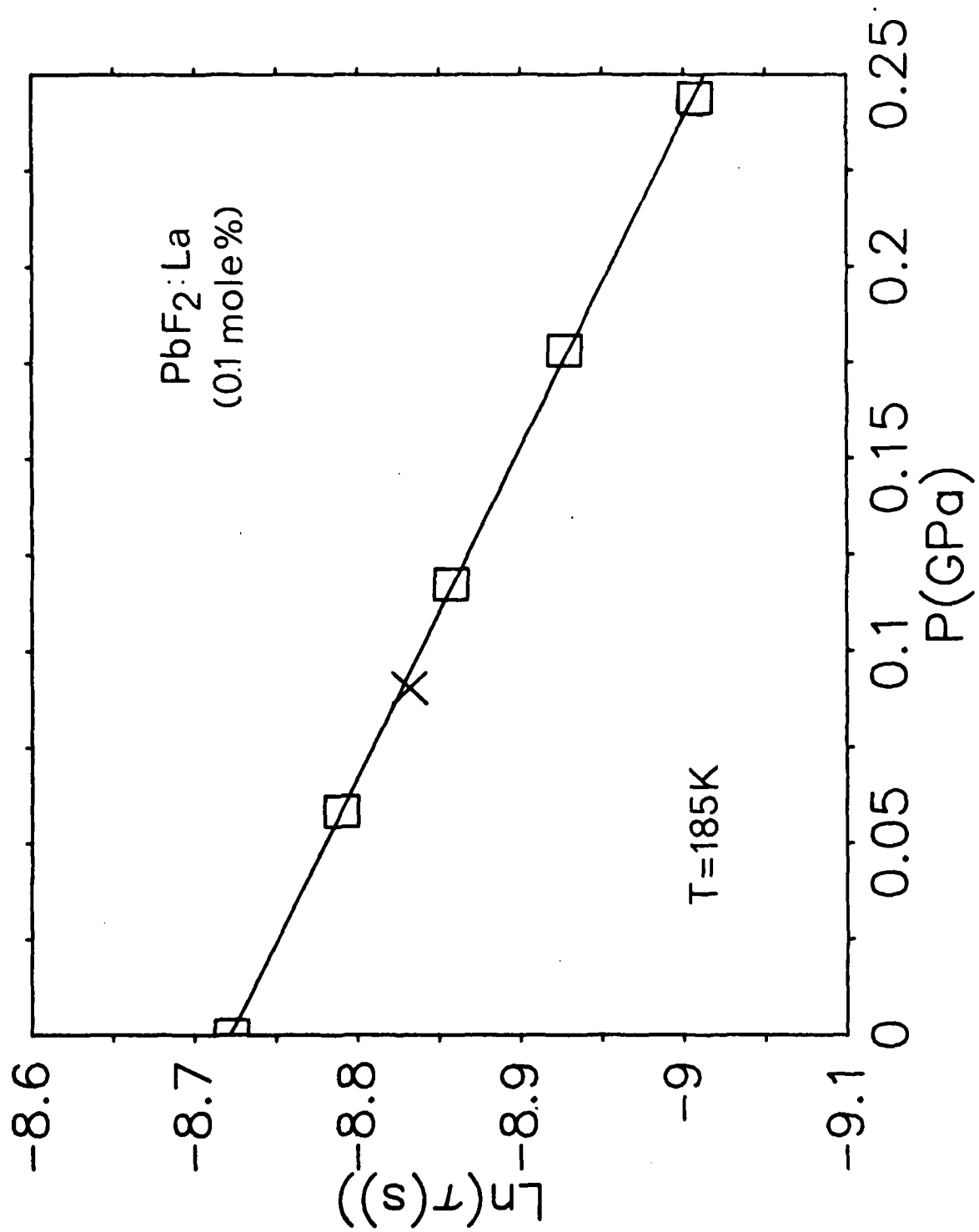
Figure 1.  $\text{Log}(\sigma T(\text{K}/\text{ohm-cm}))$  vs.  $1000/T$  ( $\text{K}^{-1}$ ) for "pure"  $\text{PbF}_2$  and 0.1 mol%  $\text{PbF}_2:\text{La}$ . The data for the pure sample is at 100 Hz and the frequencies for  $\text{PbF}_2:\text{La}$  are equal logarithmic intervals between those indicated on the drawing. Also shown is the best fit curve for  $\text{PbF}_2:\text{La}$ .

Figure 2.  $\text{Log}(f(\text{Hz}))$  vs.  $G/\omega$  (pF) for 0.1 mol%  $\text{PbF}_2:\text{La}$  at 185K and  $P=0.0001$  GPa (o) and  $P=0.244$  GPa (x). Also shown are the best fit Cole-Cole curves.

Figure 3.  $\text{Ln}(\tau(\text{s}))$  vs.  $P(\text{GPa})$  for 0.1 mol%  $\text{PbF}_2:\text{La}$ . Decreasing pressure-o; Increasing pressure-x.









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